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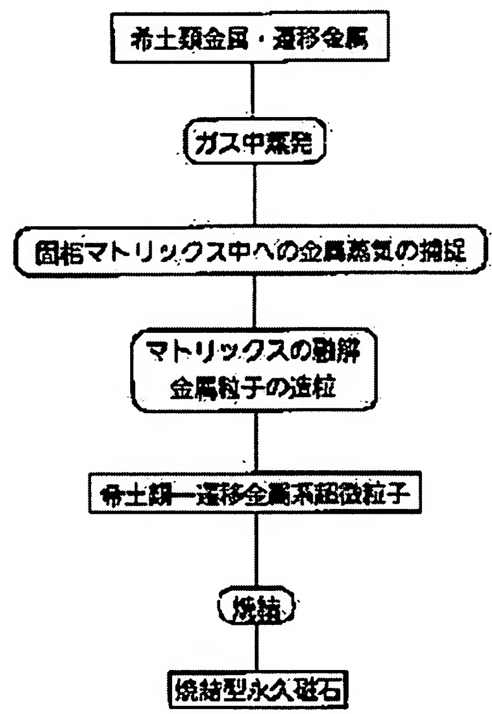
(54) PRODUCTION OF SUPERFINE PARTICLE OF RARE EARTH-TRANSITION METAL BASED COMPOUND AND APPLICATION THEREOF

(57)Abstract:

PURPOSE: To obtain superfine particle of a rare earth-transition metal based compound useful as a raw material of a high performance sintered permanent magnet by heating and vaporizing a rare earth metal and a transition metal in an atmosphere of ammonia or the like to make a corresponding nitride or the like and after condensing the vapor, melting and flocculating the matrix.

CONSTITUTION: The superfine particle of rare earth-transition metal based compound having several tens nm particle size or below and represented Ln-Fe- N base, Ln-Fe-C base, Ln-Fe-B base (Ln is rare earth element) or the like is produced in accordance with a producing process expressed by the figure by the combination of vaporization-in-gas method and matrix isolation method. A prescribed quantity of rare earth metal and transition metal are mounted in a vacuum chamber capable of introducing a gas and provided with electrodes. A condensable gas such as ammonia is introduced after evacuating and simultaneously is cooled by liquefied nitrogen or the like to condense the gas and to make a solid phase matrix. Next, each metal is vaporized by heating and isolated and frozen into the matrix. Then, after a brine is removed, the superfine particle is obtained by melting and coagulating.



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CLAIMS

[Claim(s)]
[Claim 1] The technique of manufacturing the rare earth-transition-metals system compound ultrafine particle represented by **, such as a Ln-Fe-N system which has the grain size of dozens of nm or less, a Ln-Fe-C system, and a Ln-Fe-B system (Ln: rare earth elements), by the approach which combined gas evaporation and the matrix isolating method.

[Translation done.]

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1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] Being able to manufacture the mixture of each component used as the rare earth-transition-metals system intermetallic compound which has outstanding magnetic properties by this invention, or its raw material with the gestalt of an ultrafine particle dozens of nm or less, the obtained particle powder has an application as a high performance sintering mold permanent magnet.
[0002]
[Description of the Prior Art] The conventional atomization to a rare earth-transition-metals system intermetallic compound is performed by the approach of evaporating in gas the approach and each component metal which grind mechanically the intermetallic compound produced by the metallurgical approach.
[0003]
[The technical problem that invention will be solved] the magnitude of the particle obtained with the conventional atomization technique -- at most -- it is submicron extent and the ultrafine particle with a high degree of sintering with a still smaller particle size is not obtained yet an old place.
[0004]
[Means for Solving the Problem] In order to attain the aforementioned object, the rare earth metal and transition metals used as a raw material Ammonia, The compound steam which the shape of an atom was made to carry out heating evaporation once, and this generated in ambient atmospheres of **, such as methane and diboron hexahydride, in a suitable solid phase matrix After a isolation supplement, Development of the manufacture process made to condense mutually is effective in the dissolved matrix, and in this invention, the ultrafine particle of 10nm or less of particle-size numbers is manufactured, and it is characterized by producing a high performance sintering mold permanent magnet, using this as a raw material.
[0005]
[Function] By this invention, production can manufacture the rare earth-transition-metals system compound ultrafine particle which has a difficult particle size of dozens of nm or less by the conventional atomizing method.
[0006] Manufacture is performed by warming each component of the atom-like rare earth metal which carried out evaporation evaporation once in ambient atmospheres of **, such as ammonia, methane, and diboron hexahydride, and transition metals after isolation freezing, warming a matrix in a solid phase matrix, and making it condense between each component. The ultrafine particle of the particle size made into the object is producible by controlling the conditions of warming in that case.
[0007] It can consider as the ultrafine particle of a corresponding rare earth-transition-metals system nitride, carbide, or a way ghost by carrying out the heating evaporation of the metal in ammonia, methane, or diboron hexahydride especially.
[0008] Moreover, since the obtained ultrafine particle has high reactivity, it is characterized by the degree of sintering being high. Therefore, the sintering mold permanent magnet of the rare earth-transition-metals system intermetallic compound of a difficulty degree of sintering is also producible.
[0009]
[Example] According to the production process shown in drawing 1 , the compound ultrafine particle of **, such as a Ln-Fe-N system, a Ln-Fe-C system, and a Ln-Fe-B system, can be manufactured.
[0010] First, the electrode was prepared via the current installation terminal in the vacuum chamber in which gas installation is possible. This was equipped with the sample heating unit and it loaded with the rare earth metal and transition metals of the specified quantity into it. While introducing the gas of condensation nature including **, such

as after [evacuation] ammonia, and a hexane, the vacuum chamber was cooled with refrigerants, such as liquid nitrogen, and it considered as the solid phase matrix by making a vacuum chamber wall condense the above-mentioned gas. Next, the heating evaporation of each metal was carried out in ambient atmospheres of **, such as ammonia of place constant pressure introduced in the system, methane, and diboron hexahydride, and isolation freezing of the compound steam was carried out into the solid phase matrix. Then, the refrigerant was removed, the matrix was dissolved and the condensation between the frozen compound clusters was advanced. The rare earth-transition-metals system ultrafine particle with a particle size predetermined by controlling temperature and time amount has been manufactured at that time.

[0011] The ultrafine particle of a corresponding rare earth-transition-metals system nitride, carbide, and a way ghost has been manufactured by carrying out the heating evaporation of the metal in ammonia, methane, and diboron hexahydride especially.

[0012] The scanning electron microscope photograph of a Sm-Fe-N system compound ultrafine particle is shown in drawing 2 . The particle size of each particle is about 80nm or less, and showed high reactivity, such as igniting easily by friction in atmospheric air. Therefore, the hydrogen absorption property seen with the degree of sintering between particles or some intermetallic compounds also improved remarkably.

[0013] Powder X diffraction drawing of the Sm-N system produced in the ammonia ambient atmosphere to drawing 3 and a Sm-Fe-N system compound particle is shown. Although the peak which can belong to SmN was observed by the Sm-N system, the reinforcement was very weak and peak width was also broadcloth. On the other hand by the Sm-Fe-N system sample, a peak was not looked at by the diffraction diagram, but the obtained ultrafine particle was amorphous.

[0014]

[Effect of the Invention] Compared with the rare earth-transition-metals system compound particle from which this invention is manufactured by grinding of the conventional intermetallic compound, or evaporation among gas, particle size can manufacture an ultrafine particle with single or more small figures. Therefore, a high degree of sintering appears by reactant improvement in the particle itself, and effectiveness is in manufacture of the high performance sintering mold permanent magnet which was not able to be produced conventionally.

[Translation done.]

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審査請求　未請求　請求項の数1（全 3 頁）

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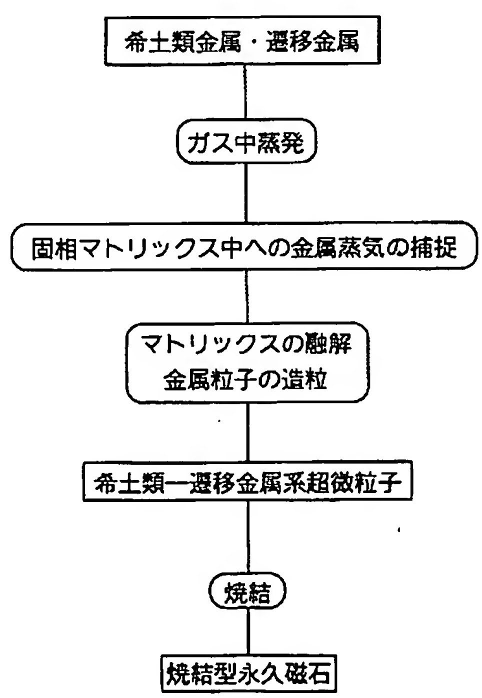
(54)【発明の名称】　希土類－遷移金属系化合物超微粒子の製造と応用

(57)【要約】

【目的】　高性能焼結型永久磁石への良好な原料となる希土類－遷移金属系金属間化合物あるいは各成分化合物の混合物からなる超微粒子（粒径数十nm以下）を、ガス中蒸発法とマトリックス単離法を組み合わせた方法で製造する。

【構成】　アンモニア、メタン、ジボラン、等々の雰囲気中で希土類金属および遷移金属を加熱気化させることにより、対応する希土類－遷移金属系窒化物、炭化物あるいはほう化物とし、この蒸気を真空室内に凝縮させたアンモニア、ブタン、等々の固相マトリックスで単離補足する。次に、マトリックスを融解させることにより各化合物間で凝集させ、粒径数十nm以下の希土類－遷移金属系化合物超微粒子を製造する。さらに、得られた超微粒子は高性能焼結型永久磁石の原料として使用される。



【特許請求の範囲】

【請求項1】　数十nm以下の粒子サイズを有するLn－Fe－N系、Ln－Fe－C系、Ln－Fe－B系（Ln：希土類元素）、等々に代表される希土類－遷移金属系化合物超微粒子を、ガス中蒸発法とマトリックス単離法を組み合わせた方法により製造する技術。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明により、優れた磁気特性を有する希土類－遷移金属系金属間化合物あるいはその原料となる各成分の混合物を数十nm以下の超微粒子の形態で製造でき、得られた粒子粉末は高性能焼結型永久磁石としての用途をもつ。

【0002】

【従来の技術】希土類－遷移金属系金属間化合物に対する従来の微粒子化は、冶金学的方法で作製した金属間化合物を機械的に粉砕する方法および各成分金属をガス中で蒸発させる方法により行なわれている。

【0003】

【発明が解決しようという課題】従来の微粒子化技術で得られる粒子の大きさは高々サブミクロン程度であり、さらに粒径が小さく焼結性が高い超微粒子はこれまでのところまだ得られていない。

【0004】

【課題を解決するための手段】前記の目的を達成するためには、原料となる希土類金属および遷移金属をアンモニア、メタン、ジボラン、等々の雰囲気中で一度原子状に加熱気化させ、これにより生成した化合物蒸気を適当な固相マトリックス中で単離補足後、融解させたマトリックス中で相互に凝集させる製造プロセスの開発が有効であり、本発明では粒径数十nm以下の超微粒子を製造し、これを原料として用い高性能焼結型永久磁石を作製することを特徴としている。

【0005】

【作用】本発明では、従来の微粒子化法では作製が困難であった数十nm以下の粒径を有する希土類－遷移金属系化合物超微粒子を製造することができる。

【0006】製造は、アンモニア、メタン、ジボラン、等々の雰囲気中で一度蒸発気化させた原子状希土類金属および遷移金属の各成分を固相マトリックス中に単離凍結後、マトリックスを加温し各成分間で凝集させることにより行う。その際、加温の条件を制御することにより目的とする粒径の超微粒子を作製することができる。

【0007】特に、アンモニア、メタンあるいはジボラン中で金属を加熱気化させることにより、対応する希土類－遷移金属系窒化物、炭化物あるいはほう化物の超微粒子とすることができる。

【0008】また、得られた超微粒子は高い反応性を有するため、その焼結性が高いことを特徴としている。そのため、難焼結性の希土類－遷移金属系金属間化合物の焼結型永久磁石も作製することができる。

【0009】

【実施例】図1に示す製造工程により、Ln－Fe－N系、Ln－Fe－C系、Ln－Fe－B系、等々の化合物超微粒子を製造することができる。

【0010】まず、ガス導入が可能な真空室内に電流導入端子を経由して電極を設けた。これに試料加熱部を装着し、その中に所定量の希土類金属と遷移金属を装填した。真空排気後アンモニア、ヘキサン、等々を始めとする凝縮性のガスを導入すると共に真空室を液体窒素等の冷媒で冷却し、上記のガスを真空室内壁に凝縮させることにより固相マトリックスとした。次に、系内に導入した所定圧のアンモニア、メタン、ジボラン、等々の雰囲気中でそれぞれの金属を加熱気化させ、固相マトリックス中に化合物蒸気を単離凍結した。引き続き、冷媒を取り除きマトリックスを融解させ、凍結されていた化合物クラスター間での凝集を進行させた。その際、温度と時間を制御することで所定の粒径をもつ希土類－遷移金属系超微粒子を製造できた。

【0011】特に、アンモニア、メタンおよびジボラン中で金属を加熱気化させることにより、対応する希土類－遷移金属系窒化物、炭化物およびほう化物の超微粒子を製造できた。

【0012】図2に、Sm－Fe－N系化合物超微粒子の走査型電子顕微鏡写真を示す。各微粒子の粒径はおよそ80nm以下であり、大気中での摩擦により容易に発火するなど高い反応性を示した。そのため、粒子間の焼結性や一部の金属間化合物で見られる水素吸蔵特性も著しく向上した。

【0013】図3に、アンモニア雰囲気中で作製したSm－N系およびSm－Fe－N系化合物微粒子の粉末X線回折図を示す。Sm－N系では、SmNに帰属できるピークが観測されるが、その強度は極めて弱く、またピーク幅もブロードであった。一方、Sm－Fe－N系試料では回折図にはピークが見られず、得られた超微粒子は非晶質であった。

【0014】

【発明の効果】本発明は、従来の金属間化合物の粉砕あるいはガス中蒸発で製造される希土類－遷移金属系化合物粒子と比べ、粒径が一桁以上も小さい超微粒子を製造することができる。そのため、粒子自身の反応性の向上により高い焼結性が出現し、従来作製が不可能であった高性能焼結型永久磁石の製造に効果がある。

【図面の簡単な説明】

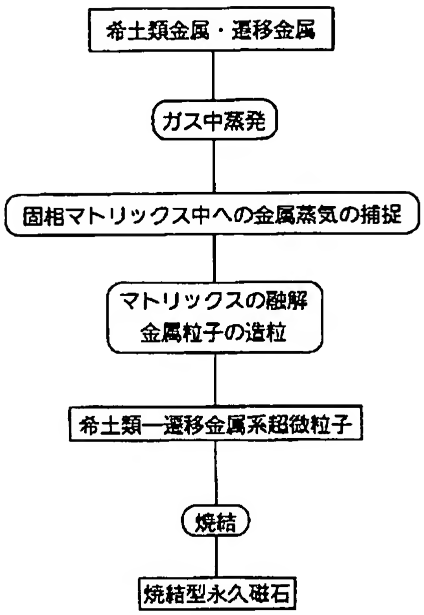
【図1】希土類－遷移金属系化合物超微粒子の製造工程図である。

【図2】Sm－Fe－N系化合物超微粒子の走査型電子顕微鏡写真である。

【図3】粉末X線回折図である。ただし、(a)はSm－N系化合物超微粒子、(b)はSm－Fe－N系化合物超

微粒子である。

【図1】



【図2】

【図3】